

Vanishing Meissner effect as a hallmark of in-plane FFLO instability in superconductor – ferromagnet layered systems

S. Mironov,¹ A. Mel'nikov,¹ and A. Buzdin²

¹*Institute for Physics of Microstructures, Russian Academy of Sciences, 603950 Nizhny Novgorod, GSP-105, Russia*

²*Institut Universitaire de France and University Bordeaux,
LOMA UMR-CNRS 5798, F-33405 Talence Cedex, France*

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We demonstrate that in a wide class of multilayered superconductor – ferromagnet structures (e.g., S/F, S/F/N and S/F/F') the vanishing Meissner effect signals the appearance of the in-plane Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) modulated superconducting phase. In contrast to the bulk superconductors the FFLO instability in these systems can emerge at temperatures close to the critical one and is effectively controlled by the S layer thickness and the angle between magnetization vectors in the F/F' bilayers. The predicted FFLO state reveals through the critical temperature oscillations vs the perpendicular magnetic field component.

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The diamagnetic supercurrent and resulting magnetic field expulsion observed in seminal experiments by Walther Meissner [1] are known to be one of fundamental phenomena peculiar to superconducting materials. The London theory [2] gives us a famous expression for the supercurrent density $\mathbf{j} = -e^2 n_s \mathbf{A} / mc$ originating from the phase rigidity of the wave function of superconducting electrons. Here n_s is the density of superconducting electrons, m is the electron mass and \mathbf{A} is the vector potential. Assuming naturally the electron density and mass to be positive we always get the $\mathbf{j} - \mathbf{A}$ relation corresponding to a diamagnetic response. Recently this observation has been questioned in several theoretical works [3–5] predicting the sign change in the London relation and an unusual paramagnetic response of the hybrid superconductor/ferromagnet (S/F) and superconductor/normal metal (S/N) systems. Such anomalous Meissner effect has been attributed to the odd-frequency spin-triplet superconducting correlations generated due to proximity effect [6].

For S/F systems the inversed sign of the Meissner currents is closely related to the oscillatory behavior of the Cooper pair wave function inside the ferromagnet [7, 8]. These oscillations are known to cause a number of important fingerprints of the S/F proximity effect including local increase in the electronic density of states at the Fermi energy [9–12], π -Josephson junction formation [13, 14] and non-monotonic dependencies of the critical temperature of S/F bilayers on the F layer thickness [15, 16].

The unusual electromagnetic response contribution becomes even stronger for a superconductor placed in contact with a composite F/F' layer with different mutual orientations of the magnetic moments. Such systems are known to reveal so-called long-range triplet superconducting correlations predicted in Refs. [3, 17]. The local supercurrent density can be written as $\mathbf{j} = -e^2(n_s - n_t)\mathbf{A}/mc$, where $n_s(n_t)$ is the density of the singlet (triplet) condensate. Different character of the n_s

and n_t components decay leads to the change in the sign of the local response, i.e., inversion of the Meissner effect. During the last two years an important breakthrough in the experimental observation of the long-ranged triplet proximity effect occurred [18, 19]. All this makes very timely the study of the magnetic response of the proximity induced triplet superconductivity. Note that the first experimental measurements [20] of the London penetration depth in thin S/F bilayers revealed a slightly non-monotonic dependence of the penetration depth on the F layer thickness, which was in accordance with the theoretical analysis [21].

In this paper we address the intriguing problem of the Meissner response of the S/F systems exhibiting the above sign change in the relation between the supercurrent density and vector potential and show that the anomalous Meissner effect can cause the in-plane Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) instability [22, 23] of the superconducting uniform state. To elucidate our main results we start from rather general arguments illustrating the physical origin of the instability in systems with the anomalous Meissner effect. Considering the local supercurrent density ($\mathbf{j} = -\delta F_A / \delta \mathbf{A} = -e^2 n_s \mathbf{A} / mc$) as a variational derivative of the free energy functional we find the corresponding free energy term: $F_A = \int (e^2 n_s \mathbf{A}^2 / 2mc) dV$. The sign change in the current – vector potential relation can be considered as a change in the sign of the effective mass. Introducing the superconducting order parameter phase φ and writing the free energy in the gauge-invariant form

$$F_A = \int \frac{e^2 n_s}{2mc} \left(\mathbf{A} - \frac{\Phi_0}{2\pi} \nabla \varphi \right)^2 dV, \quad (1)$$

where Φ_0 is the flux quantum, one can clearly see that the negative local effective mass can, in principle, result in the instability of the homogeneous state with $\varphi = const$, $\mathbf{A} = 0$ and appearance of the phase φ modulation. Namely such a situation is realized at the transi-

tion to the non-uniform FFLO state (see the discussion, for example, in [24]). As a consequence, the above expression describing the linear current response should be reconsidered for a new inhomogeneous ground state.

To illustrate the above general arguments by a concrete example of instability we hereafter focus on the consideration of thin film structures of total thickness much smaller than the screening length. This assumption allows us to consider only the currents flowing in the film plane and neglect the change of the vector potential on the structure thickness. Introducing the in-plane FFLO modulation vector \mathbf{k} so that $\varphi = \mathbf{k}\mathbf{r}_{\parallel}$ we find:

$$F_A = \left(\mathbf{A}_{\parallel} - \frac{\Phi_0}{2\pi} \mathbf{k} \right)^2 S \int \frac{e^2 n_s}{2mc} dx, \quad (2)$$

where the x axis is chosen perpendicular to the film plane, S is the sample area in the (yz) plane, \mathbf{A}_{\parallel} and \mathbf{r}_{\parallel} are parallel to the film. All the states with $\Lambda^{-1} = \int (e^2 n_s / 2mc) dx < 0$ are clearly unstable and, thus, the boundary of the in-plane FFLO instability is given by the condition $\Lambda^{-1} = 0$ of vanishing Meissner effect for the in-plane field. Note that the above arguments, being applied for the FFLO state itself, clearly show that in the modulated state the Meissner response should be diamagnetic. Thus, in the systems under consideration the paramagnetic Meissner response appears to be impossible.

We now proceed with the microscopic calculations of the FFLO critical temperature and magnetic screening length for three particular structures (S/F, S/F/N and S/F/F') shown in Fig. 1. Note that for S/F bilayers the modulated along the F layer state has been suggested in Ref. [25] but later it has been pointed out [26] that the conclusions of Ref. [25] are based on the wrong boundary conditions assuming the modulation of the order parameter only in the F layer. In contrast with Ref. [25] in our case the same modulation is present both in S and F layers. Somewhat similar non-uniform phase has been predicted for a ferromagnetic cylinder covered by the superconducting shell [27]. Interestingly in ^3He films the non-uniform superfluid p-wave state may be stimulated by the surface scattering of quasiparticles [28]. In our

calculations we assume that: (i) the system is in a dirty limit; (ii) the exchange field h in the ferromagnet is much larger than the critical temperature T_{c0} of the isolated S layer; (iii) the thickness of the S layer d_s is smaller than the coherence length $\xi_s = \sqrt{D_s/2\pi T}$ (D_s is the diffusion constant in a superconductor), so we can neglect the variation of the order parameter function Δ across the S layer; (iv) all interfaces are transparent.

Near the critical temperature the anomalous Green function

$$\hat{f} = \begin{pmatrix} f_{11} & f_{12} \\ f_{21} & f_{22} \end{pmatrix} = (f_s + \mathbf{f}_t \hat{\sigma}) i \hat{\sigma}_y. \quad (3)$$

satisfies the linearized Usadel equation [30]

$$\frac{D}{2} \nabla^2 \hat{f} - \omega_n \hat{f} - \frac{i}{2} (\mathbf{h} \hat{\sigma} \hat{f} + \hat{f} \mathbf{h} \hat{\sigma}) + \hat{\Delta} = 0, \quad (4)$$

where $\hat{\Delta} = \Delta i \hat{\sigma}_y$ is the superconducting gap function, $\omega_n = \pi T(2n+1)$ are the Matsubara frequencies, and D is the diffusion constant, which may be different for different layers. In the absence of the barriers between layers the function \hat{f} as well as the combination $\sigma \partial_x \hat{f}$ are continuous at each interface (σ is the Drude conductivity of the corresponding layer). We assume Fermi velocities in all layers to be equal, so that the ratio between conductivities of different layers is the same as the ratio between the corresponding diffusion constants. The critical temperature T_c of the system is determined by the component f_{12}^S of the Green function in the superconductor in accordance with the self-consistency equation

$$\Delta \ln \frac{T_c}{T_{c0}} + \sum_{n=-\infty}^{\infty} \left(\frac{\Delta}{|2n+1|} - \pi T_c f_{12}^S \right) = 0, \quad (5)$$

where T_{c0} is the critical temperature of the isolated superconducting film.

In the limit of weak screening the Meissner response averaged over the structure thickness d_0 takes the form

$$\lambda^{-2} = \frac{1}{\Lambda d_0} = \frac{16\pi^3 T_c}{ec\Phi_0 d_0} \sum_{n=0}^{\infty} \int \sigma (|f_s|^2 - |\mathbf{f}_t|^2) dx. \quad (6)$$

This expression clearly shows that the triplet component provides the negative contribution to λ^{-2} . To describe the FFLO state we assume the gap $\Delta(\mathbf{r}_{\parallel}) = \Delta_0 \exp(i\mathbf{k}\mathbf{r}_{\parallel})$ and the anomalous Green function $\hat{f} = \hat{\varphi}(x) \exp(i\mathbf{k}\mathbf{r}_{\parallel})$ to be spatially modulated.

We start from the simplest case of a bilayer (see Fig. 1(a)), which consists of a thin S film and F layer of the thickness $d_f \ll \xi_n$. The exchange field \mathbf{h} in the F layer is uniform and directed along the z -axis, so that $f_{11} = f_{22} = 0$. Substituting the modulated Green function into the Usadel equation and solving it under the assumption, that the function \hat{f} weakly varies across the S layer, we obtain the components $f_{12}^{S(F)}$ in the S(F) layer

$$f_{12}^S = \frac{\Delta_0 e^{i\mathbf{k}\mathbf{r}_{\parallel}}}{\omega_n + \tau_s^{-1}(k)}, \quad f_{12}^F = f_{12}^S \frac{\cosh(q_k(x - d_f))}{\cosh(q_k d_f)}, \quad (7)$$

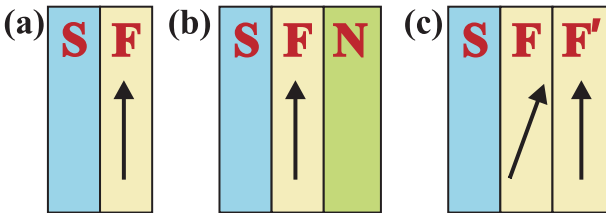


FIG. 1: (Color online) The sketch of the hybrid structures under consideration. S layer is placed in contact with (a) F film, (b) F/N bilayer and (c) F/F' bilayer with different magnetic moment orientations shown by arrows.

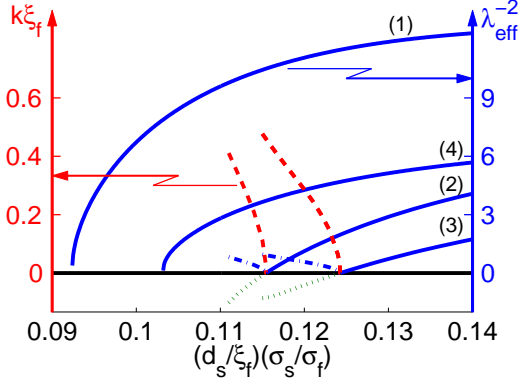


FIG. 2: (Color online) Magnetic screening parameter λ^{-2} (blue solid lines) in the uniform superconducting state and the optimal FFLO modulation vector k_0 (red dashed curves) vs the S layer thickness d_s for the S/F bilayer. Blue dash-dotted curves drawn by hand illustrate the behavior of the magnetic screening parameter in the FFLO regime while green dotted curves correspond to the λ^{-2} behavior calculated for the unstable uniform state. We take $\xi_{s0} = \sqrt{D_s/4\pi T_{c0}} = 0.1\xi_f$ and (1) $d_f = 0.75\xi_f$, (2) $d_f = 1.0\xi_f$, (3) $d_f = 1.2\xi_f$, (4) $d_f = 2.0\xi_f$. Also we denote $\lambda_{eff}^{-2} = \lambda^{-2} (T_c(0)ec\Phi_0 d_0/2\pi\sigma_s d_s \Delta^2)$.

where $q_k = \sqrt{q^2 + k^2}$, $q = (1+i)/\xi_f$, and $\xi_f = \sqrt{D_f/h}$ is the coherence length in the ferromagnet. The complex pair-breaking parameter

$$\tau_s^{-1}(k) = \frac{D_s}{2}k^2 + \frac{D_s}{2d_s} \frac{\sigma_f}{\sigma_s} q_k \tanh(q_k d_f) \quad (8)$$

determines the critical temperature $T_c(k)$ of the S film:

$$\ln \frac{T_c(k)}{T_{c0}} = \Psi\left(\frac{1}{2}\right) - \text{Re}\Psi\left(\frac{1}{2} + \frac{\tau_s^{-1}(k)}{2\pi T_c(k)}\right), \quad (9)$$

where Ψ is the Digamma function. Note that these results can be obtained by replacing $\omega_n \rightarrow \omega_n + D_{s(f)}k^2/2$ in the Usadel equation for the uniform state.

The effective magnetic screening length in the uniform state can be expressed through the derivative of the above expression for T_c at $k = 0$:

$$\lambda^{-2} = -\frac{d_s \sigma_s \Delta^2 D_s}{2ec\Phi_0 d_0 T_c(0)} \left[1 - \text{Re} \left\{ \nu \Psi_1 \left(\frac{1}{2} + \nu \right) \right\} \right] \frac{\partial T_c}{\partial k^2} \Big|_{k=0}. \quad (10)$$

Here $\nu = \tau_s^{-1}(0)/2\pi T_c(0)$ and Ψ_1 is the trigamma function. Calculating the derivative of the Eq. (9) we find the result obtained in [21]. The condition of the stability of the uniform superconducting state, $\partial T_c / \partial k^2 (k=0) < 0$, imposes a diamagnetic character of the Meissner response for the magnetic field parallel to the plane of the layers.

For $d_f \sim \xi_f$ the contribution from the F layer to the Meissner response coefficient λ^{-2} can become negative. For S/F bilayers with a large difference in the diffusion constants ($D_f/D_s \gg h/T_{c0}$) the screening parameter λ^{-2} can even vanish at some critical thickness

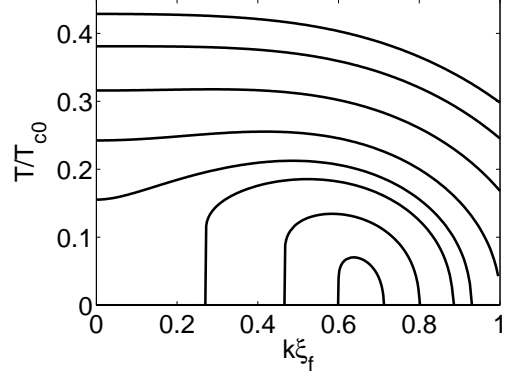


FIG. 3: The dependencies of the critical temperature T_c vs the modulation vector k for different thicknesses d_s of the S film. We take $\xi_{s0} = \sqrt{D_s/4\pi T_{c0}} = 0.1\xi_f$, $d_f = 1.2\xi_f$ and the following set of values $(d_s/\xi_f)(\sigma_s/\sigma_f)$: 0.13, 0.125, 0.12, 0.1165, 0.1148, 0.114, 0.113, 0.1125. The increasing d_s thickness corresponds to the increasing T_c maximum.

$d_{sc} \sim (\sigma_f/\sigma_s)\xi_f$. At the critical thickness $d_s = d_{sc}$ the derivative $\partial T_c / \partial (k^2)|_{k=0}$ turns to zero and $d_s < d_{sc}$ the superconducting transition occurs not to the uniform but to the modulated FFLO state with the modulation vector $k_0 \neq 0$. The typical dependencies $\lambda^{-2}(d_s)$ are shown by blue solid curves in Fig. 2. The dependencies $k_0(d_s)$ for different d_f are shown by red dashed curves in Fig. 2. The corresponding dependencies $T_c(k)$ are shown in Fig. 3. It is interesting that the FFLO state can survive even for parameter range corresponding to a complete suppression of the uniform BCS state for all temperatures.

Discussing the physical reason of the FFLO phase emerging in S/F bilayer we should note that the proximity effect with a ferromagnet plays a role of the pair-breaking effect at the S/F interface. The FFLO-like modulation of the pairing wave function weakens such pair-breaking, but at the same time this modulation suppresses partially the critical temperature of the S layer. The efficiency of the first (second) mechanism is governed by the diffusion coefficient $D_f(D_s)$ and finally for $D_f/D_s \gg h/T_{c0}$ the critical temperature of the FFLO state may exceed that of the uniform one.

Our analysis reveals a direct relation between the vanishing Meissner effect and the FFLO phase formation. In Fig. 4 we show the distribution of the triplet and singlet components over the bilayer thickness in the BCS state close to the threshold of the FFLO instability. One can see that the triplet component providing the anomalous contribution to the Meissner effect strongly exceeds the singlet one at the free surface of the F layer. This circumstance gives a hint how to stabilize the FFLO phase: one should add the normal metal (N) layer on the top of the ferromagnetic layer (see Fig. 1(b)). Moreover such modified system allows to overcome the strong damping of T_c in the FFLO state of the S/F bilayer and get the

FFLO state for temperatures close to T_{c0} . Details of calculations can be found in Supplemental Material [29].

The appearance of the FFLO state can be effectively controlled provided we consider S/F/F' structures (see Fig.1(c)) with a certain angle θ between the magnetization vectors in the F and F' layers. Such systems are recently discussed as possible candidates for spin valve devices [31–33]. For non-collinear magnetic moments the triplet component of the anomalous Green function generated in the F film becomes long-range in the F' layer and decays at a distance of the order of $\xi_n \gg \xi_f$ (where $\xi_n = \sqrt{D_{f'}/4\pi T_{c0}}$) while the singlet component is fully damped at a distance $\sim \xi_{f'} = \sqrt{D_{f'}/\hbar}$ from the F/F' interface. As a result, if the thickness $d_{f'}$ of the F' layer strongly exceeds ξ_f then the corresponding contribution into the screening parameter λ^{-2} is *always* negative and can become comparable with the one from the S film. In the simplest case of small (large) thickness of the F(F') layer, i.e. $d_f \ll \xi_f$ and $d_{f'} \rightarrow \infty$, the long-ranged triplet component $f_{11}^{F'}$ in the F' layer is proportional to $(d_f/\xi_f)^2 \sin \theta$. For large diffusion constant $D_{f'}$, the ratio between the negative contribution coming from the F' layer and positive S layer contribution to the screening parameter λ^{-2} can become of the order of unity for

$$\sin^2 \theta \gtrsim \frac{D_s d_s}{D_{f'} \xi_n} \left(\frac{\xi_f}{d_f} \right)^4. \quad (11)$$

Varying the angle θ one can trigger the transition from the uniform state, realized for θ close to zero and π , to the FFLO state, which is favorable for θ close to $\pi/2$. Thus, the formation of the FFLO phase should affect the angular dependence of the critical temperature in S/F/F' spin valves devices.

Experimentally the appearance of the FFLO state can be identified by the observation of the critical temperature oscillations vs magnetic field H perpendicular to the plane of the layers [34]. For simplicity we consider here only the case of a S/F bilayer. Choosing an appropriate

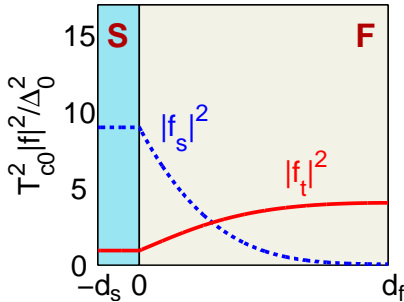


FIG. 4: (Color online) The spatial profile of the singlet (blue dashed curve) and triplet (red solid curve) components of the anomalous Green function in the S/F bilayer at $T = T_c(0)$ and $\omega_n = \pi T$. We take $\xi_{s0} = 0.1\xi_f$, $d_f = 1.2\xi_f$ and $(d_s/\xi_f)(\sigma_s/\sigma_f) = 0.13$.

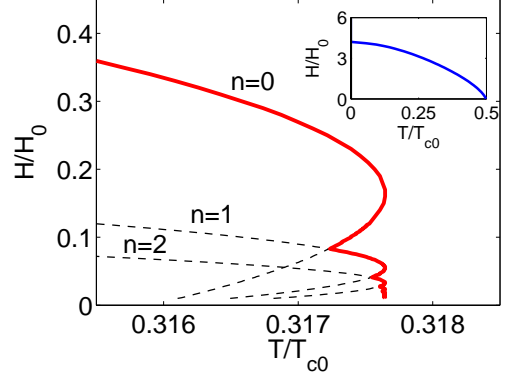


FIG. 5: (Color online) The phase diagram of the S/F bilayer in the FFLO regime (red curve). Dashed curves correspond to dependencies $T_c(H)$ for different n . We put here $\xi_{s0} = 0.1\xi_f$, $d_f = 1.2\xi_f$ and $d_s = 0.12(\sigma_f/\sigma_s)\xi_f$. For comparison in the inset we show the H-T phase diagram for $d_s = 0.14(\sigma_f/\sigma_s)\xi_f$ corresponding to the uniform superconducting state. We denote $H_0 = \Phi_0/4\pi\xi_f^2$.

vector potential $\mathbf{A}(\mathbf{r}_{\parallel})$ in the plane of the layers we get the Usadel equation for the component f_{12} in the form

$$\frac{D}{2} \left[\partial_x^2 + \left(\partial_{\mathbf{r}_{\parallel}} - \frac{2\pi}{\Phi_0} \mathbf{A}(\mathbf{r}_{\parallel}) \right)^2 \right] f_{12} - (\omega_n + ih) f_{12} + \Delta = 0. \quad (12)$$

The solution of the Eq. (12) takes the form: $f_{12} = \chi_n(\mathbf{r}_{\parallel}) \varphi(x)$, where $\chi_n(\mathbf{r}_{\parallel})$ is an eigenfunction of the Hamiltonian $\hat{H} = -[\partial_{\mathbf{r}_{\parallel}} - 2\pi\mathbf{A}(\mathbf{r}_{\parallel})/\Phi_0]^2$. The critical temperature corresponding to the n -th Landau level is defined by Eq. (9) with $k^2 \rightarrow 2\pi H(2n+1)/\Phi_0$. The competition between levels with different n results in a peculiar dependence $T_c(H)$ shown in Fig. 4.

In conclusion, we predict that vanishing Meissner effect in thin-film multilayered S/F systems should result in the in-plane FFLO instability which is particularly important for designing the π -junction or spin valve devices. Interestingly that in contrast with the original FFLO phase [22, 23] which emerges at relatively low temperature, the modulated phase in S/F/N or S/F/F' heterostructures may appear near the critical temperature of S layers. The appearance of the FFLO phase besides the anomalous behavior of the screening, should also result in the oscillatory-like temperature dependence of the perpendicular critical field. For the S layer thickness exceeding the screening length the FFLO instability can not, of course, expand into the bulk superconductor. In this case the surface instability can reveal through the formation of the vortex sheet parallel (and positioned close) to the S/F interface suppressing, thus, the anomalous part of the Meissner response. The linear vorticity density should be proportional to the FFLO modulation vector k_0 . Note in conclusion that similar instabilities could also appear for the superconductors with anisotropic pairing

where the paramagnetic currents are caused by the surface – induced Andreev bound states [35, 36].

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